

Single wall carbon nanotube double quantum dot

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We report on two top-gate defined, coupled quantum dots in a semiconducting single wall carbon nanotube, constituting a tunable double quantum dot system. The single wall carbon nanotubes are contacted by titanium electrodes, and gated by three narrow top-gate electrodes as well as a back-gate. We show that a bias spectroscopy plot on just one of the two quantum dots can be used to extract the addition energy of both quantum dots. Furthermore, honeycomb charge stability diagrams are analyzed by an electrostatic capacitor model that includes cross capacitances, and we extract the coupling energy of the double quantum dot.

Electronic transport in single quantum dots (QDs) defined in single wall carbon nanotubes (SWCNTs) has been studied intensively over the last decade.^{1,2,3} These devices are typically made by placing metal electrodes directly on top of a SWCNT resulting in tunnel barriers at each SWCNT-metal interface, and gated by using the substrate as one global gate. Recent studies have shown that it is possible to locally gate and locally deplete a small segment of a SWCNT.^{4,5} By placing several such local gates on top of a SWCNT, a double quantum dot (DQD) with tunable inter-dot coupling can be made.^{6,7,8,9,10} A DQD is a desirable system since it can be used in the field of quantum computation as e.g. a single charge qubit or two interacting spin qubits.¹¹ The advantage of making DQDs in SWCNTs instead of other material systems such as GaAs/AlGaAs is that SWCNTs are thought to have a longer spin decoherence time. An important source of decoherence is the hyperfine coupling between the electron in the QD and the nuclei. ¹²C is the abundant isotope in natural carbon and has no net nuclear spin. The hyperfine coupling will therefore be highly reduced in SWCNTs.

In this Letter we present a fabrication scheme to contact and place three narrow local gates on top of a SWCNT. We show that a device fabricated by this method can be used to define two coupled QDs in series. The addition energies of both QDs are estimated from a low temperature bias spectroscopy plot on just one of the QDs. These addition energies are then used together with a honeycomb charge stability diagram to estimate the electrostatic coupling energy of the DQD.

The devices are made on a highly doped silicon substrate capped by a 0.5 μm thermally oxidized SiO_2 layer, and we use the substrate as a back-gate to tune the global potential of the SWCNT. A set of alignment marks are made by electron beam lithography (EBL), which are used to accurately position the following three steps of EBL. First, islands of catalyst material consisting of a suspension of aluminum oxide nanoparticles in methanol with dissolved iron nitrate and molybdenum acetate are placed at specific positions, see Fig. 1(a). For easy liftoff and an even distribution of the catalyst we use a thick double layered resist (9% copolymer, and 4% PMMA) and spin on the liquid catalyst at 1000 rpm for 150 s. The

SWCNTs are then grown by chemical vapor deposition from the catalyst islands in a ceramic tube furnace at $\sim 900^\circ\text{C}$ with a controlled flow of gasses, Ar: 1.1 L/min, H_2 : 0.1 L/min, and CH_4 : 0.5 L/min.^{12,13} Typically only a few or one SWCNT will grow several μm away from the island, see Fig. 1(b). The alignment marks are secondly used to position source and drain electrodes consisting of 50 nm titanium with a separation of 1.8 μm . Since the SWCNTs tend to bundle together into ropes within a distance of about $\sim 1 \mu\text{m}$ from the island, the electrode nearest to the island are positioned $\sim 2 \mu\text{m}$ from island, thus favoring contact to long straight SWCNTs. In about 30% of our devices only one tube is contacted. Third, three narrow gate electrodes are defined by EBL using a thin double layer resist (6% copolymer, and 2% PMMA) and positioned between the source and drain electrodes, by use of the alignment marks. The gates consist of five evaporations of aluminum each 2 nm thick and oxidized in air for about 1 min, and a top layer of titanium. We contact the EBL-structures with a final step of optical lithography to be able to bond the device onto a chip-carrier.

The fabrication scheme presented here has good possibilities to be scaled up to produce several devices in each batch. In Fig. 1(a) we show a pattern with four potential devices. Several of these patterns could easily be made in each batch, where we currently make just two. An atomic force microscope (AFM) micrograph of a finished device is shown in Fig. 1(b) where only one SWCNT is contacted. The three gates are named G1, CG (center gate), and G2 starting from the source electrode. We apply source-drain voltage (V_{sd}) to the source electrode and keep the drain electrode at ground. The nanotube in the device that we present measurements on in this Letter has a height (diameter) measured with an AFM of about $\sim 1 \text{ nm}$. It shows an ambipolar characteristic at room temperature as seen in Fig. 1(c), which suggests that it is a small band gap semiconducting SWCNT. We can thus use the back-gate to tune the global potential of the device from electron to hole transport. In the rest of the letter the measurements are made through the valence band with a back-gate voltage of $V_{BG} = -6 \text{ V}$, to ensure that transport is governed by holes.

Figure 2(a) shows a bias spectroscopy plot at 300 mK

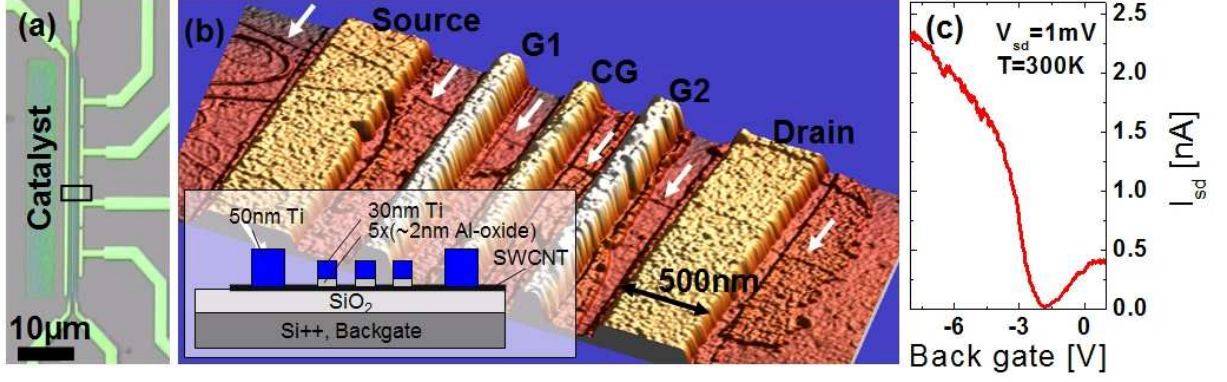


FIG. 1: (Color) (a) Optical image of 4 potential devices consisting of one common source electrode, three common top-gate electrodes, and four individual drain electrodes. On the left hand side of the source electrode an island of catalyst material is positioned from where the carbon nanotubes grow. (b) Atomic force microscope micrograph of the region indicated by the black rectangle in (a). To the left (close to the catalyst island) several tubes can be seen, but only one tube has grown several μm away from the island (indicated with white arrows). Source and drain electrodes consisting of 50 nm titanium, and three top-gate electrodes consisting of five 2 nm layers of air-oxidized aluminum and 30 nm titanium, are positioned directly on top of the tube. Some resist residue can be seen around some of the leads. Insert: Schematic side view of the device. (c) Current through the device as function of voltage applied to the back-gate at room temperature, and with 1 mV source-drain voltage.

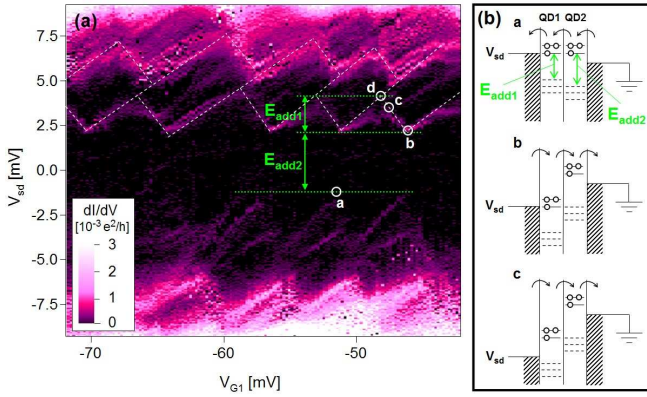


FIG. 2: (Color) (a) Bias spectroscopy plot of differential conductance (dI/dV) versus source-drain voltage and voltage applied to G1, with $V_{CG} = 0\text{ V}$, $V_{G2} = 1.1\text{ V}$, and $V_{BG} = -6\text{ V}$ at 300 mK. The white dashed lines are guidelines to the eye, indicating charge degeneracy lines. The addition energies of each quantum dot are indicated with green arrows. (b) Schematic figures of the hole transport through the double quantum dot at positions indicated with letters in (a). Solid and dashed lines are filled and empty hole states, respectively. The coupling energy is here neglected since it is much smaller than the addition energies (see below).

of the differential conductance versus V_{sd} and voltage applied to G1 (V_{G1}), with CG and G2 kept constant at $V_{CG} = 0\text{ V}$, and $V_{G2} = 1.1\text{ V}$, respectively. That is, in Fig. 2(a) QD1 is probed by the source electrode from the left hand side, and a discrete energy level of QD2 from the right hand side. Around zero bias the device does not conduct, and the onset of conductance

is asymmetric around zero bias. The onsets of conductance at point **a** and **b** in Fig. 2(a) are positioned at $V_{sd} \sim -1.5\text{ mV}$, and $V_{sd} \sim 2.1\text{ mV}$, respectively. The conductance gap is constant in the bias spectrum in Fig. 2(a), and also constant in the whole gate range that we measured ($V_{G1} = \pm 100\text{ mV}$). This gap in conductance is due to the DQD nature of the device, where the first QD (QD1) is tuned by G1, and the second QD (QD2) is tuned by G2. Both QDs have Coulomb blockade (CB) oscillations and since QD2 is in CB for the chosen gate voltage on G2, transport is blocked whenever the bias is smaller than the addition energy of QD2. Since QD1 is probed from the right hand side by energy levels from QD2 and because the chemical potential of the drain lead is aligned asymmetrically between two successive energy levels of QD2, the conductance gap is asymmetric around zero bias (see Fig. 2(b)). At point **a** the energy levels of the two QDs are aligned with the chemical potential of the source lead, and we have hole transport from drain to source. From point **a** to point **b** the energy level of QD1 and the chemical potential of the source lead are kept aligned and shifted together, while QD2 is kept constant in CB, i.e., no sequential tunneling is possible. At point **b** the chemical potential of the source lead and the energy level of QD1 becomes aligned with the next energy level of QD2, which gives hole transport from source to drain. The conductance gap is therefore a measure of the addition energy of QD2 (E_{add2}).

Above and below the conductance gap structures similar to CB diamonds for a single QD are observed. These structures are due to CB in QD1 and illustrated from point **b** to point **d** through point **c** in Fig. 2(a). Along the line from point **b** to point **d** the ground level in each QD are kept aligned, while the chemical potential of the

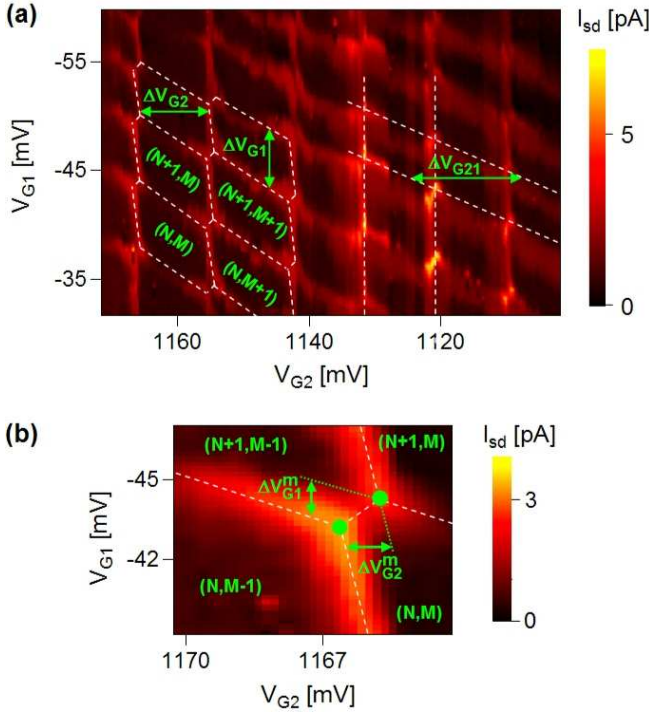


FIG. 3: (Color) Charge stability diagrams at 300 mK of the measured current as function of V_{G1} , and V_{G2} , with $V_{CG} = 0$ V, $V_{BG} = -6$ V, and $V_{sd} = 2$ mV. (a) Honeycomb pattern with relative number of holes in each QD indicated with green numbers. The white dashed lines are guidelines to the eye. (b) Close-up of one set of triple points at the position indicated with the relative hole numbers.

source electrode is shifted downwards to align with the next energy level of QD1. Because of the capacitive coupling between source and QD1 a negative compensating gate voltage on G1 is needed to keep the ground levels in QD1 and QD2 aligned. The distance from point **b** to point **d** in source-drain voltage is therefore a measure of the addition energy of QD1 (E_{add1}). Since no odd/even or four-period structures originating from the level spacings in the two dots is observed in either bias spectroscopy plots or honeycomb charge stability diagrams (see below), we estimate the level spacings to be much smaller than the charging energies. The two methods to read-off the addition energy of QD1 and QD2 gives on average $E_{add1} \sim 2.2$ meV, and $E_{add2} \sim 3.6$ meV. At higher bias (above the level of point **d**) more structures are observed. A thorough explanation of these structures is beyond the scope of this letter but an interesting subject for further study.

Figure 3 shows a charge stability diagram of current through the DQD as function V_{G1} , and V_{G2} . Honeycomb structures can be identified throughout the plot which is a clear sign of a DQD with inter-dot coupling. Within each honeycomb structure the number of holes in each QD is constant, as indicated with relative hole numbers (N, M) in Fig. 3(a) and (b). At the corners of these hon-

eycombs so-called triple points are located, where three charge states are degenerate, e.g., (N, M) , $(N+1, M)$, and $(N, M+1)$. At these triple points an increase in current is observed consistent with sequential tunneling becoming possible via the three degenerate charge states. Furthermore, the overall slope of the honeycombs as illustrated with white dashed lines in the right side of Fig. 3(a) can be used to estimate cross capacitances. When G2 is decreased by ΔV_{G21} (indicated in Fig. 3(a)) one hole is added to QD1, i.e., a cross capacitance from G2 to QD1 exists. Since the vertical distance (ΔV_{G12}) between the two almost vertical lines to the right in Fig. 3(a) tends to infinity, there are almost zero cross capacitance from G1 to QD2.

The observed splitting of adjacent triple points, as shown in Fig. 3(b) is due to coupling between the QDs. The electrostatic coupling energy (E_{Cm}) is defined as the change in potential energy of QD1 when a hole is added to QD2, or vice versa. We have extended the electrostatic capacitor model in Ref. [14] to include cross capacitances. We find that the electrostatic coupling energy can be given in terms of quantities directly observable in a bias spectroscopy plot and in a honeycomb charge stability diagram:

$$E_{Cm} = E_{add1(2)} \cdot \frac{\Delta V_{G1(2)}^m}{\Delta V_{G1(2)}} \cdot \frac{\Delta V_{G12(21)}}{\Delta V_{G12(21)} - \Delta V_{G1(2)}^m} \quad (1)$$

where $\Delta V_{G1(2)}$, and $\Delta V_{G1(2)}^m$, is the size of the honeycombs and the splitting of the triple points, as illustrated in Fig. 3(a) and (b), respectively. The last term in Eq. (1) accounts for the cross capacitances and goes to unity when there are no cross capacitances, i.e., $\Delta V_{G12(21)}$ goes to infinity. An average estimate of $\Delta V_{G1(2)}$ from all the honeycombs seen in Fig. 3(a) gives $\Delta V_{G1(2)} \sim 6(10)$ mV. The estimated values of $\Delta V_{G1(2)}^m$ and $\Delta V_{G12(21)}$ is; $\Delta V_{G1(2)}^m \sim 1.25(1.10)$ mV, and $\Delta V_{G21} \sim 20$ mV (ΔV_{G12} tends to infinity). Since the gate voltages used in the bias spectrum in Fig. 2 and the gate voltages used in the charge stability diagram in Fig. 3 are chosen to be roughly the same, we can use the addition energies found above in the estimation of E_{Cm} . From these experimental values two consistent estimates of the electrostatic coupling energy is obtained, $E_{Cm} \sim 0.46(0.42)$ meV.

In conclusion we have presented a fabrication scheme that in each batch can produce several devices for electronic transport in a SWCNT with three narrow top-gates. We show that a device fabricated by this method can be used to define two coupled QDs in series. From a bias spectroscopy plot of just one of the QDs the addition energies of both QDs are extracted. Furthermore, an electrostatic capacitor model that includes an observed cross capacitance is used on a honeycomb charge stability diagram to extract two consistent estimates of the coupling energy.

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- ¹ S. J. Tans, M. H. Devoret, H. Dai, A. Thess, R. E. Smalley, L. J. Geerling, and C. Dekker, *Nature (London)* **386**, 474 (1997).
- ² J. Nygård, D. Cobden, and P. E. Lindelof, *Nature* **408**, 342 (2000).
- ³ W. Liang, M. Bockrath, D. Bozovic, J. H. Hafner, M. Tinkham, and H. Park, *Nature (London)* **411**, 665 (2001).
- ⁴ T. W. Tombler, C. Zhou, J. Kong, and H. Dai, *Appl. Phys. Lett.* **76**, 2412 (2000).
- ⁵ M. J. Biercuk, N. Mason, and C. M. Marcus, *Nano Lett.* **4**, 1 (2004).
- ⁶ N. Mason, M. J. Biercuk, and C. M. Marcus, *Science* **303**, 655 (2004).
- ⁷ M. J. Biercuk, S. Garaj, N. Mason, J. M. Chow, and C. M. Marcus, *Nano Lett.* **5**, 1267 (2005).
- ⁸ S. Sapmaz, C. Meyer, P. Beliczynski, P. Jarillo-Herrero, and L. P. Kouwenhoven, *Nano Lett.* **6**, 1350 (2006).
- ⁹ M. R. Gräber, W. A. Coish, C. Hoffmann, M. Weiss, J. Furer, S. Oberholzer, D. Loss, and C. Schönenberger, *Phys. Rev. B* **74**, 075427 (2006).
- ¹⁰ M. R. Gräber, M. Weiss, and C. Schönenberger, *Semicond Sci. Technol.* **21**, 64-68 (2006).
- ¹¹ D. Loss, and D. P. DiVincenzo, *Phys. Rev. A* **57**, 120 (1998).
- ¹² J. Kong, H. T. Soh, A. M. Cassell, C. F. Quate, and H. Dai, *Nature* **395**, 878 (1998).
- ¹³ K. Grove-Rasmussen, H. I. Jørgensen, and P. E. Lindelof, *cond-mat/0601371* (2006).
- ¹⁴ W. G. van der Wiel, S. De Franceschi, J. M. Elzerman, T. Fujisawa, and L. P. Kouwenhoven, *Rev. Mod. Phys.* **75**, 1 (2003).